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# The impacts of the activity coefficient on heating and evaporation of ethanol/gasoline fuel blends

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## Highlights

- Effects of the activity coefficient on blended fuel evaporation
- Effects of ethanol and gasoline fuel blends on droplet lifetimes
- The evolution of droplet surface temperatures and radii

## Abstract

The evolutions of droplet radii and temperatures for ethanol and gasoline fuels and their blends are investigated using a modified version of the Discrete Component (DC) model, taking into account the effect of the activity coefficient (AC). The universal quasi-chemical functional-group AC (UNIFAC) model is used to predict the ACs of the blended ethanol and gasoline fuels approximated by 21 components. In contrast to previous studies, it is shown that droplet lifetimes predicted for pure gasoline are not always shorter than those predicted for ethanol/gasoline blends. They depend on the total vapour pressure of the mixture. It is shown that the original DC model predicts ethanol/gasoline fuel droplet lifetimes with errors up to 5.7% compared to those predicted using the same model but with the ACs obtained from the UNIFAC model.

**Keywords:** Activity coefficient, Ethanol, Evaporation, Fuel blends, Gasoline, Heating

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## 1. Introduction

Heating and evaporation of droplets are important processes in numerous applications, including those in internal combustion engines [1,2], spray coating [3], fire suppression [4], the pharmaceutical industry [5], and agriculture [6,7]. This has stimulated intensive research to develop robust models for the description of these processes [1,8–10].

Our analysis is focused primarily on ethanol/gasoline fuel blend droplets, the interest in which has been mainly stimulated by the depletion of fossil fuels and environmental concerns. The heating and evaporation of these blends have been investigated numerically and experimentally [11–14]. In these studies, however, gasoline has been approximated by iso-octane or a mixture of iso-octane/n-heptane, whilst the commercial gasoline fuels consist of tens of hydrocarbons [15]. The effects of fuel compositions, transient diffusion of species, temperature gradient, and recirculation inside moving droplets on their heating and evaporation have been commonly described using the Discrete Component (DC) model [9,16], and the Effective Thermal Conductivity/Effective Diffusivity (ETC/ED) model [8]. These models have been validated against experimental data [17–19].

The DC model was used previously for the analysis of blended fuel droplet heating and evaporation, including blends of diesel/biodiesel and ethanol/gasoline fuels [17,20–24]. In these studies, however, Raoult's law was assumed to be valid (the activity coefficient (AC) was assumed equal to one). Unlike fossil fuels, ethanol and biodiesel fuels are polar liquids. Therefore, Raoult's law may not be suitable for predicting the vapour pressures of these fuel blends [25]. To address this issue, in our analysis we took into account the contributions of non-unity ACs. In some studies (e.g. [26]), the Wilson equation was used for the predictions of ACs. The Wilson equation is a simple approach, but limited to binary components. In the general case, the universal quasi-chemical functional-group AC (UNIFAC) model is believed to be the most suitable for prediction of the multi-component ACs [11,27]. In [28], the UNIFAC model was used to predict the ACs of 20 components in gasoline FACE C and 98 components in diesel fuel. This approach, however, was based on the initial molar fractions of components and droplet surface temperatures. In the current analysis, we investigate the impact of transient ACs on the evolutions of blended ethanol/gasoline fuel droplet temperatures and radii. The transient droplet surface temperatures and diffusion of 21 components are taken into account using the UNIFAC model. The governing equations and main features of the DC model used in our analysis are summarised in [8,29], and will not be discussed in this paper. The main features of the model and the implementation of UNIFAC equations into relevant equations of the DC model are described in Section 2. The results predicted by the modified DC model, using the corrected ACs, are presented and discussed in Section 3. The main results are summarised in Section 4.

## **2. The model**

The DC model used in our analysis is based on the analytical solutions to the heat transfer and species diffusion equations inside droplets [9]. The effects of recirculation on species diffusion and heat conduction inside droplets are taken into account, using the Effective Thermal Conductivity/Effective Diffusivity (ETC/ED) model [30]. The evaporation rate of a droplet is described by the following equation:

$$\dot{m}_d = -2\pi R_d D_v \rho_{\text{total}} B_M \text{Sh}_{\text{iso}}, \quad (1)$$

where  $D_v$  is the binary diffusion coefficient of vapour in gas (air),  $\rho_{\text{total}} = \rho_g + \rho_v$  is the total density of the mixture of vapour and ambient gas (assumed not to depend on the distance from the droplet surface),  $\text{Sh}_{\text{iso}}$  is the Sherwood number for isolated droplets (taking into account the effects of droplet motion and evaporation),  $B_M = \frac{Y_{vs} - Y_{\infty}}{1 - Y_{vs}}$  is the Spalding mass transfer number, and  $Y_{vs}$  and  $Y_{\infty}$  are vapour mass fractions in the vicinity of the droplet surface and in the far-field, respectively,  $Y_{vs} = \sum_i Y_{vis}$  and  $Y_{vis}$  are the mass fractions of individual species  $i$ , calculated from the vapour molar fractions at the droplet surface ( $X_{vis}$ )[16]:

$$X_{vis} = \gamma_i \frac{X_{lis} p_{vis}^*}{p}, \quad (2)$$

where  $p$  is the total (ambient) pressure,  $X_{lis}$  is the liquid molar fraction of the  $i^{\text{th}}$  species at the surface of the droplet,  $\gamma_i$  is the AC of the  $i^{\text{th}}$  species, and  $p_{vis}^*$  is the saturated pressure of the  $i^{\text{th}}$  species in the absence of other species.

In contrast to [10,17,20,24,31–33], where Raoult's law is assumed to be valid ( $\gamma_i = 1$ ), our study accounts for the values of ACs taking into consideration the effect of corrected partial pressures of vapour components. The UNIFAC model is used for the estimation of the ACs of 21 components of ethanol/gasoline fuel blends. This model includes two terms: the combinatorial term (C) and residual term (R), taking into account the contribution of the excess entropy and the effect of the excess enthalpy, respectively. The excess entropy is inferred from various shapes and sizes of molecules or functional groups of atoms (hereafter referred to as groups, see Appendix A), while the excess enthalpy is inferred from interactions between molecules or groups [27,34]. The UNIFAC equation for the AC of component  $i$  in a multi-component mixture is presented as [35]:

$$\ln \gamma_i = \ln \gamma_i^C + \ln \gamma_i^R \quad (3)$$

where

$\ln \gamma_i^C = \ln \frac{\Phi_i}{X_i} + \frac{Z}{2} q_i \ln \frac{\theta_i}{\Phi_i} + l_i - \frac{\Phi_i}{X_i} \sum_j X_j l_j$  is the combinatorial part,  $\ln \gamma_i^R = \sum_k v_k^i (\ln \Gamma_k - \ln \Gamma_k^i)$  is the residual part,  $l_i = \frac{Z}{2} (r_i - q_i) - (r_i - 1)$ ,  $Z = 10$ ,  $\theta_i = \frac{q_i X_i}{\sum_j q_j X_j}$  is the area fraction of each molecule in the mixture,  $\Phi_i = \frac{r_i X_i}{\sum_j r_j X_j}$  is the segment (volume) fraction of each molecule,  $r_i = \sum_k v_k^i R_k$  is the volume parameter,  $q_i = \sum_k v_k^i Q_k$  is the surface parameter,  $\ln \Gamma_k = Q_k \left[ 1 - \ln(\sum_m \theta_m \psi_{mk}) - \sum_m \frac{\theta_m \psi_{km}}{\sum_n \theta_n \psi_{nm}} \right]$ ,  $\theta_m = \frac{Q_m X_m}{\sum_n Q_n X_n}$  is the area fraction of group  $m$  (see Appendix A),  $X_i$  is the molar fraction of liquid component  $i$  (the same as  $X_{lis}$  in Equation (2)),  $X_m$  is the molar fraction of group  $m$ , and  $R_k$  and  $Q_k$  are the van der Waals volumes and surface areas for each group, respectively [34,35].  $R_k$  is the volume occupied by each group in the molecule, while  $Q_k$  is the surface area occupied by each group in the molecule. Both  $R_k$  and  $Q_k$  are functions of bond distances, bond angles, contact distances, and shapes

that are characteristic of the group [36]. The values of  $R_k$  and  $Q_k$  are presented in Table 4, Appendix A.  $v_k^i$  is the number of groups in molecule  $i$ .  $\Gamma_k$  is the residual AC of group  $k$  in the mixture and  $\Gamma_k^i$  is the residual AC of group  $k$  in a reference solution containing only molecules of type  $i$  (for example  $\Gamma_k^i$  for the  $\text{CH}_2\text{OH}$  group in ethanol refers to a solution containing 50%  $\text{CH}_2\text{OH}$  and 50%  $\text{CH}_3$ , while  $\text{CH}_2\text{OH}$  in 1-pentanol refers to a solution of 20%  $\text{CH}_2\text{OH}$ , 60%  $\text{CH}_2$ , and 20%  $\text{CH}_3$ ). In other words,  $\Gamma_k^i$  deals with each group in each molecule, while  $\Gamma_k$  deals with each group in the mixture.  $\psi_{mn} = e^{-\left(\frac{a_{mn}}{T}\right)}$  is the interaction and temperature dependent coefficient,  $a_{mn}$  is the group-interaction parameter between groups  $m$  and  $n$  (see Table 5, Appendix A) [35],  $m$  and  $n$  refer to the group in the mixture (in the case of  $\Gamma_k$ ) or in the molecule (in the case of  $\Gamma_k^i$ ).  $T$  is the interface temperature.

The liquid properties are calculated at the average temperature inside droplets, while the gas properties are calculated at the reference temperature  $T_r = \frac{2}{3}T_s + \frac{1}{3}T_g$ , where  $T_s$  and  $T_g$  are the droplet surface and ambient gas temperatures, respectively. Enthalpy of evaporation and saturated vapour pressure are estimated at  $T_s$ .

### 3. Results

The total vapour pressures versus molar fractions of ethanol/gasoline in the liquid phase (indicated as EMX, where X is the percentage of ethanol in the mixture) at 296 K and 350 K are presented in Figure 1. In this figure, a comparison between the two approaches, Raoult's and UNIFAC, is shown. In Raoult's law, the AC is equal to unity, while in the UNIFAC model, the values of multi-component ACs are used.

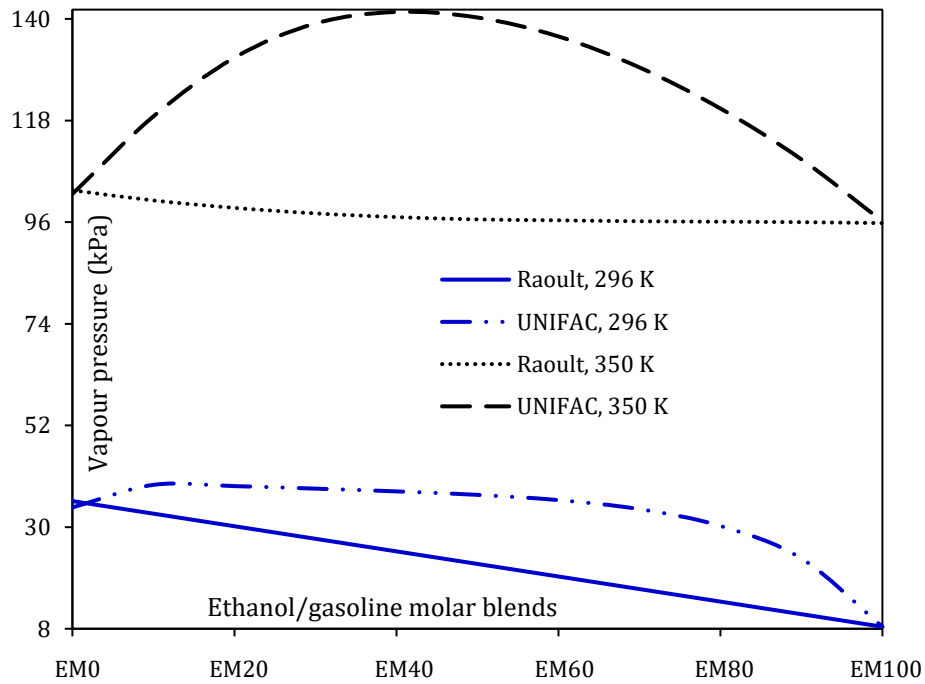
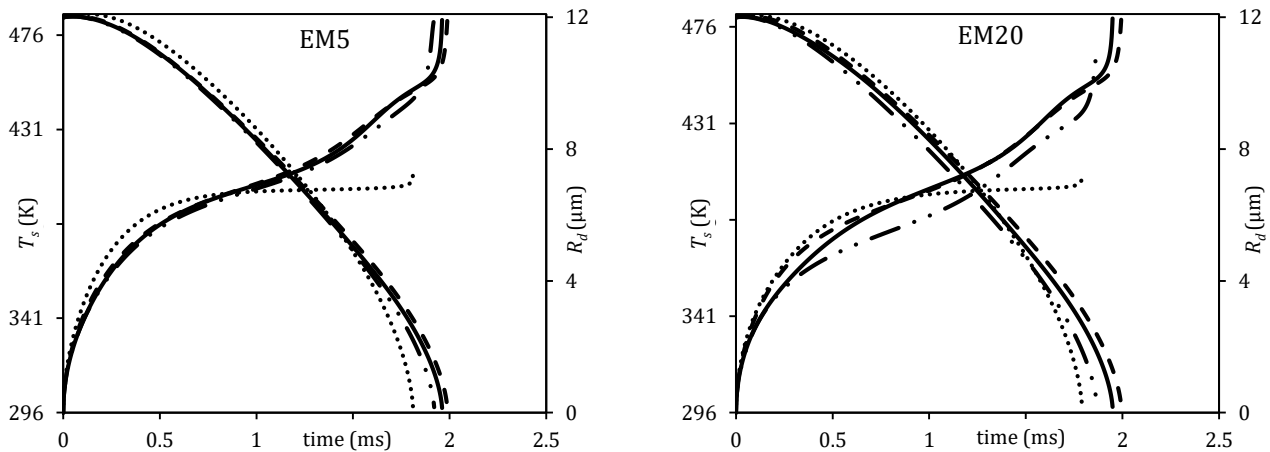


Figure 1. Total vapour pressure of various ethanol/gasoline molar blends (EM0–EM100), predicted by Raoult's law and the UNIFAC model at  $T = 296\text{K}$  and  $350\text{ K}$ .

As can be seen from this figure, the multi-component ACs have significant impact on the predictions of the total vapour pressure of ethanol/gasoline blends. For low ethanol molar fractions, breaks in the hydrogen bonds lead to a reduction in the inter-molecular forces [14]. This leads to an increase in the total vapour pressure. For high ethanol molar fractions, however, this pressure decreases as the contribution of the hydrogen bonds becomes important [14]. These predictions agree with those inferred from the experimental results presented in [37–39] (see Appendix B).

The heating and evaporation of blended ethanol/gasoline fuel droplets was investigated using the same operating conditions as in [17,32] but taking into account the impacts of non-unity ACs. The initial droplet radius was taken equal to  $R_{do} = 12 \mu m$ , and its constant velocity in still air and initial temperature were assumed equal to  $U_d = 24$  m/s and  $T_{do} = 296$  K, respectively. The ambient air pressure and temperature were assumed constant and equal to  $p_g = 9$  bar and  $T_g = 545$  K, respectively.

The evolutions of droplet surface temperatures  $T_s$  and radii  $R_d$  versus time for various initial ethanol molar fractions (EM85, EM50, EM20 and EM5) are shown in Figure 2. The mixtures EM85, EM50, EM20 and EM5 are approximately equivalent to the volume fractions 70% ethanol/30% gasoline, 29% ethanol/71% gasoline, 9% ethanol/91% gasoline, and 2% ethanol/98% gasoline, respectively. In this figure, the results predicted by four models are compared. The first one (labelled ‘Ethanol/iso-octane’) is based on the transient UNIFAC model to predict the ACs for a binary mixture, in which gasoline fuel is approximated with iso-octane. The second one (labelled ‘Raoult’) is based on the assumption that the AC is equal to one, taking into account the full composition (20 components) of gasoline fuel. The third model (labelled ‘Steady UNIFAC’) is based on the steady-state UNIFAC model, in which the ACs are predicted based on the initial liquid fuel composition (21 components of gasoline and ethanol fuels) and temperature. In the fourth model (labelled ‘Transient UNIFAC’), the ACs are calculated based on the surface compositions and temperatures at each time-step.



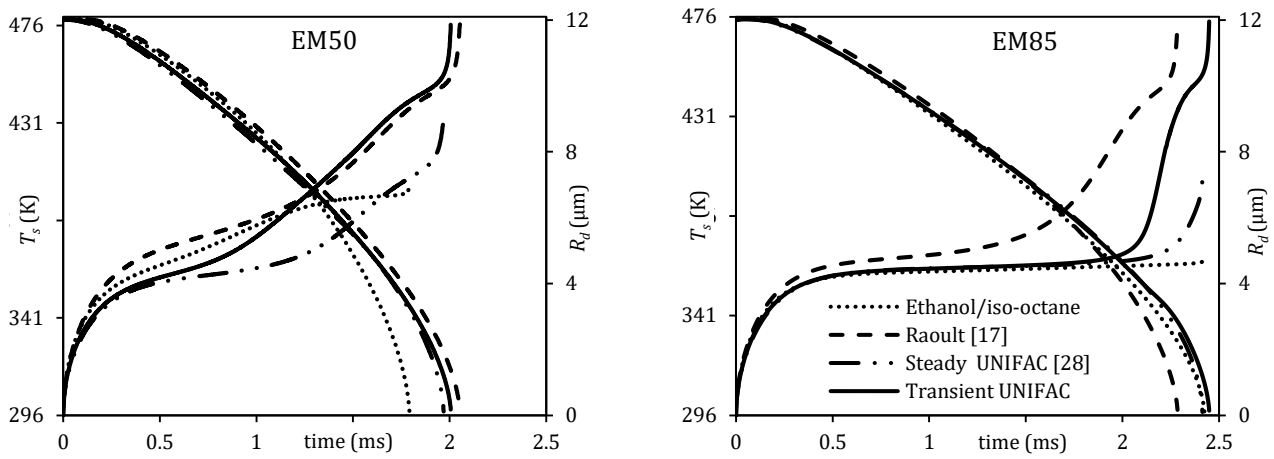


Figure 2. Droplet surface temperatures and radii versus time for ethanol/gasoline blends for various approaches to calculating the total vapour pressures at droplet surfaces.

As can be seen in Figure 2, the predicted droplet lifetimes and surface temperatures strongly depend on the type of model used to estimate the ACs. The approximation of gasoline by a single component (iso-octane) leads to significant under-predictions of droplet surface temperatures and lifetimes. These under-predictions can be as much as 22.6% and 10.7% for droplet surface temperatures and lifetimes, respectively. The predictions of droplet lifetimes using the steady-state UNIFAC model show reasonable agreement with those predicted using the transient UNIFAC model (ACs are calculated using the molar fractions of components and droplet surface temperatures at each time step). The latter two models, however, predict rather different droplet surface temperatures. Using the steady-state UNIFAC model can lead to about 14.7% error in the prediction of droplet surface temperatures, compared to those predicted using the transient approach. This is related to high dependency of the ACs on the liquid molar fractions at the surface of the droplet. The assumption of unity ACs leads to over/under-predicted ethanol/gasoline droplet lifetimes by up to 6.7%, compared with those predicted using the transient UNIFAC model. A significant impact of ACs on droplet heating and evaporation can be attributed to the fact that ethanol and gasoline fuels form blends which are far from ideal due to the high polarity of ethanol [25,37]. Hence, Raoult's law is not recommended for predicting the vapour pressures of such blends.

The droplet lifetimes and surface temperatures for various volume fractions of ethanol/gasoline blends and their differences compared to E0 (pure gasoline), taking into account the multi-component ACs, are presented in Table 1. The following volume fractions of the components were considered: E0, E5, E20, E30 E50 and E85 (EX refers to a mixture of X% volume fraction of ethanol and (100-X) % volume fraction of gasoline).

Table 1. The impact of ethanol/gasoline fuel blends on estimated droplet lifetimes and surface temperatures ( $T_s$ ) taking into account transient multi-component ACs ( $\text{Diff}\% = \frac{|\text{lifetime}, T_s \text{ Blend} - \text{lifetime}, T_s \text{ E0}|}{\text{lifetime}, T_s \text{ E0}} \times 100$ ).

blend	lifetime (ms)	Diff %	$T_s$	Diff %
E0	1.968	-	483.7	-
E5	1.954	0.71	482.0	0.35
E20	1.964	0.20	478.2	1.14
E30	2.031	3.20	476.0	1.59
E50	2.241	13.9	475.5	1.70
E85	2.563	30.2	471.4	2.54

As can be seen from Table 1, in contrast to [19], the droplet lifetimes of E5 and E20 are shorter than those of E0. This is attributed to the higher total vapour pressure of the mixture (calculated using the transient ACs) than predicted in [19]. In Tables 2 and 3, the results of droplet lifetimes and surface temperatures, obtained using Raoult's law and the transient UNIFAC model for E0–E85 blends, are compared. As these tables show, the errors in calculating droplet lifetimes and surface temperatures, using Raoult's law, can reach up to 5.7% and 0.4%, respectively.

Table 2. The estimated errors in prediction of droplet lifetimes based on Raoult's law compared to the case where the transient UNIFAC approach is used ( $\text{error}\% = \frac{|\text{lifetime}_{\text{UNIFAC}} - \text{lifetime}_{\text{Raoult}}|}{\text{lifetime}_{\text{UNIFAC}}} \times 100$ ).

blend	lifetime (ms),	lifetime (ms),	error %
	UNIFAC	Raoult	
E0	1.968	1.988	1.02
E5	1.954	1.989	1.79
E20	1.964	2.021	2.90
E30	2.031	2.058	1.33
E50	2.241	2.156	3.79
E85	2.563	2.418	5.66

Table 3. The estimated errors in prediction of droplet surface temperatures based on Raoult's law compared to the case where the transient UNIFAC approach is used ( $\text{error}\% = \frac{|T_{s \text{ UNIFAC}} - T_{s \text{ Raoult}}|}{T_{s \text{ UNIFAC}}} \times 100$ ).

blend	$T_s$ , UNIFAC	$T_s$ , Raoult	error %
E0	483.7	483.2	0.10
E5	482.0	481.5	0.11
E20	478.2	478.2	0.00
E30	476.0	476.8	0.17
E50	475.5	473.6	0.40
E85	471.4	473.1	0.36



## 4. Conclusion

The heating and evaporation of ethanol/gasoline fuel blend droplets are investigated using the discrete component model, based on the analytical solutions to the heat and mass transfer equations and the effective thermal conductivity/effective diffusivity model. The universal quasi-chemical functional-group AC (UNIFAC) model is used to predict the activity coefficients (ACs) of the components of blended ethanol/gasoline (21 components) fuels. It is found that the droplet lifetimes for the blends are not always shorter compared with those for pure gasoline droplets.

It is shown that the application of the model using Raoult's law ( $AC=1$ ) can lead to up to 5.7% errors in estimated droplet lifetimes for ethanol/gasoline blends, compared to the predictions of the same model using the transient UNIFAC approach.

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## Appendix A. UNIFAC group specifications

The molecular van der Waals volumes  $R_k$  and surface areas  $Q_k$  for various groups in gasoline and ethanol components are presented in Table 4 [35].

Table 4. Van der Waals volumes ( $R_k$ ) and surface areas ( $Q_k$ ) for various molecules and atoms.

Name	Group	Group number	$R_k$	$Q_k$
Alkanes	CH <sub>3</sub>	1	0.9011	0.848
	CH <sub>2</sub>	1	0.6744	0.540
	CH	1	0.4469	0.228
	C	1	0.2195	0.000
Olefin	CH <sub>2</sub> =CH	2	1.3454	1.176
Benzene	ACH	3	0.5313	0.400
Alkylbenzenes	ACCH <sub>3</sub>	4	1.2663	0.968
	ACCH <sub>2</sub>	4	1.0396	0.660
	ACCH	4	0.8121	0.348
Ethanol	OH	5	1.0000	1.200

As can be seen from Table 4, there are 5 groups in gasoline and ethanol fuels and each group interacts with the other 4 groups. The interaction parameters between these groups are presented in Table 5 [35].

Table 5. UNIFAC  $m$ -group and  $n$ -group interaction parameters ( $a_{mn}$ ) in K.

Group number	$n=1$	2	3	4	5
$m=1$	0.0	86.02	61.13	76.50	986.5
2	-35.36	0.0	38.81	74.15	524.1
3	-11.12	3.446	0.0	167.0	636.1
4	-69.70	-113.6	-146.8	0.0	803.2
5	156.4	457.0	89.6	25.82	0.0

## Appendix B. Validation of the predicted total ethanol/gasoline vapour pressure

The total vapour pressure of E0 – E100 ethanol/gasoline blends, predicted using the UNIFAC model, is validated against experimental data provided in [39]. Note that the difference between the vapour pressure predicted by the UNIFAC model and the experimental results presented in [39] can be at least partly attributed to the differences between gasoline FACE C and the gasoline used in the experiments (New Zealand regular grade unleaded gasoline). Thermodynamic and transport properties of these fuels, however, were found to be reasonably close.

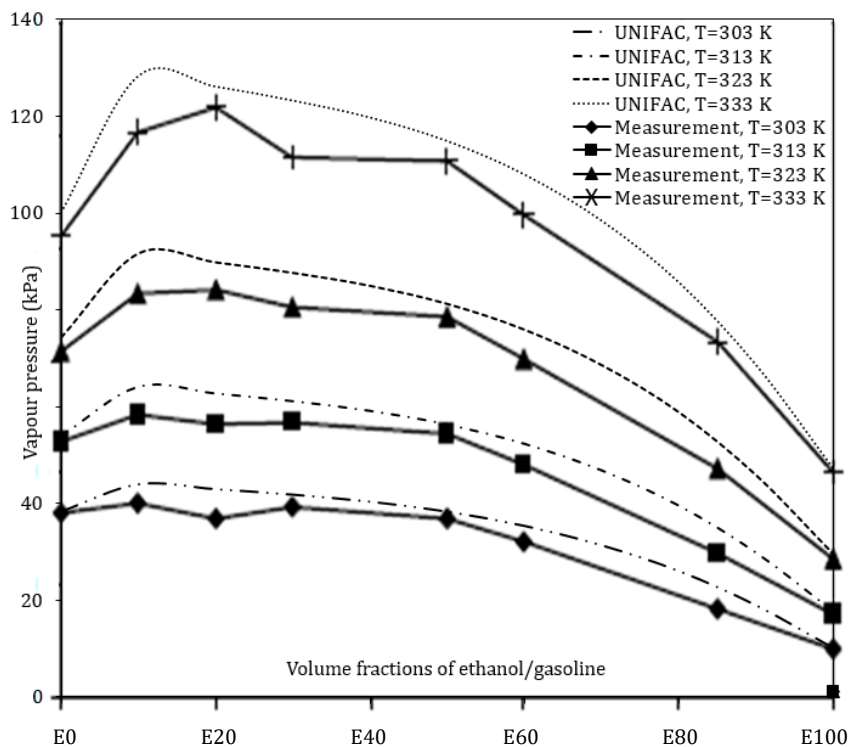


Figure 3. The predicted and experimentally observed total vapour pressures of ethanol/gasoline blends at various temperatures and ethanol volume fractions in the liquid phase.

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